



# 1. EXECUTIVE SUMMARY

## 1.1 INTRODUCTION

### 1.1.1 Purpose of the Document

The purpose of this document, Air Quality Criteria for Particulate Matter, is to present air quality criteria for particulate matter (PM) in accordance with Clean Air Act (CAA) Sections 108 and 109, which govern establishment, review, and revision of U.S. National Ambient Air Quality Standards (NAAQS).

- Section 108 directs the U.S. Environmental Protection Agency (EPA) Administrator to list pollutants that may reasonably be anticipated to endanger public health or welfare and to issue air quality criteria for them. The air quality criteria are to reflect the latest scientific information useful in indicating the kind and extent of all exposure-related effects on public health and welfare expected from the presence of the pollutant in ambient air.
- Section 109 directs the EPA Administrator to set and periodically revise, as appropriate, (a) primary NAAQS to protect against adverse health effects of listed criteria pollutants among sensitive population groups, with an adequate margin of safety, and (b) secondary NAAQS to protect against welfare effects (e.g., impacts on vegetation, crops, ecosystems, visibility, climate, man-made materials, etc.).
- To meet these CAA mandates, this document assesses the latest scientific information useful in deriving criteria as scientific bases for decisions on possible revision of current PM NAAQS. A separate EPA PM Staff Paper draws upon assessments in this document, together with other information, in delineating key information used to develop and present appropriate options for consideration by the EPA Administrator with regard to review of the PM NAAQS.

### 1.1.2 Organization of the Document

- This Executive Summary (Chapter 1) summarizes key points from ensuing chapters.
- Chapter 2 provides a general introduction, including an overview of the rationale underlying the current PM NAAQS, i.e.,  $150 \mu\text{g}/\text{m}^3$  (24-h) and  $50 \mu\text{g}/\text{m}^3$  (annual average) as  $\text{PM}_{10}$  (particles  $\leq 10 \mu\text{m}$  aerodynamic diameter,  $d_{ae}$ ).
- Chapters 3 through 7 provide background information on air quality and exposure aspects, to help to place the succeeding discussions of PM effects into perspective.
- Chapter 8 deals with visibility and climate effects; and Chapter 9 assesses materials damage, as key types of welfare effects of concern for the current PM NAAQS review. Welfare effects of PM on vegetation, crops, and ecosystems are not assessed in the document.

- Chapters 10, 11, and 12, respectively, discuss PM dosimetry, toxicology and community epidemiology information. Chapter 13 provides an integrative synthesis of key points from those health chapters and other preceding air quality and exposure chapters.

## 1.2 AIR QUALITY AND EXPOSURE ASPECTS

The document's discussion of air quality and exposure aspects considers chemistry and physics of atmospheric PM; analytical techniques for measuring PM mass, size, and chemical composition; sources of ambient PM in the United States; temporal/spatial variability and trends in ambient U.S. PM levels; and human exposure relationships.

### 1.2.1 Chemistry and Physics of Atmospheric Particles

- Airborne PM is not a single pollutant, but rather is a mixture of many subclasses of pollutants with each subclass containing many different chemical species. Atmospheric PM occurs naturally as fine-mode and coarse-mode particles that, in addition to falling into different size ranges, differ in formation mechanisms, chemical composition, sources, and exposure relationships.
- Fine-mode PM is derived from combustion material that has volatilized and then condensed to form primary PM or from precursor gases reacting in the atmosphere to form secondary PM. New fine-mode particles are formed by the nucleation of gas phase species, and grow by coagulation (existing particles combining) or condensation (gases condensing on existing particles). Fine particles are composed of (a) freshly generated particles, in an ultrafine or nuclei mode, and (b) an accumulation mode, so called because particles grow into and remain in that mode.
- Coarse-mode PM, in contrast, is formed by crushing, grinding, and abrasion of surfaces, which breaks large pieces of material into smaller pieces. They are then suspended by the wind or by anthropogenic activity. Energy considerations limit the break-up of large particles and small particle aggregates generally to a minimum size of about 1  $\mu\text{m}$  in diameter. Mining and agricultural activities are examples of anthropogenic sources of coarse-mode particles. Fungal spores, pollen, and plant and insect fragments are examples of natural bioaerosols also suspended as coarse-mode particles.
- Within atmospheric particle modes, the distribution of particle number, surface, volume, and mass by diameter is frequently approximated by lognormal distributions. Aerodynamic diameter,  $d_{ae}$ , which depends on particle density and is defined as the diameter of a particle with the same settling velocity as a spherical particle with unit density ( $1 \text{ g/cm}^3$ ) is often used to describe particle size. Typical values of the mass median aerodynamic diameter (MMAD) and geometric standard deviation ( $\sigma_g$ ) of each size mode of an aerosol are:

- Nuclei mode:	MMAD=0.05 to 0.07 $\mu\text{m}$	$\sigma_g = 1.8$
- Accumulation mode:	MMAD= 0.3 to 0.7 $\mu\text{m}$	$\sigma_g = 1.8$
- Coarse mode:	MMAD= 6 to 20 $\mu\text{m}$	$\sigma_g = 2.4$

At high relative humidities or in air containing evaporating fog or cloud droplets, the accumulation mode may be split into a droplet mode (MMAD = 0.5 to 0.8  $\mu\text{m}$ ) and a condensation mode (MMAD = 0.2 to 0.3  $\mu\text{m}$ ).

- Research studies use (a) impactors to determine mass as a function of size over a wide range and (b) particle counting devices to determine number as a function of size. Such studies indicate an atmospheric bimodal distribution of fine and coarse particle mass with a minimum in the distribution between 1 and 3  $\mu\text{m}$   $d_{ae}$ . Routine monitoring studies, however, is generally limited to measuring TSP (total suspended particles) including both fine and coarse particles up to 40 or more  $\mu\text{m}$   $d_{ae}$ ; thoracic particles or  $\text{PM}_{10}$  (upper size limited by a 50% cut at 10  $\mu\text{m}$   $d_{ae}$ ); fine particles or  $\text{PM}_{2.5}$  (upper size limited by a 50% cut point at 2.5  $\mu\text{m}$   $d_{ae}$ ), and the coarse fraction of  $\text{PM}_{10}$ , i.e., the difference between  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  ( $\text{PM}_{10-2.5}$ ). Cut points are not perfectly sharp for any of these PM indicators; some particles larger than the cutpoint are collected and some smaller-particles smaller than the cutpoint are not retained.
- The terms "fine" and "coarse" were originally intended to apply to the two major atmospheric particle distributions which overlap in the size range between 1 and 3  $\mu\text{m}$  diameter. Now, fine has come to be often associated with the  $\text{PM}_{2.5}$  fraction and coarse is often used to refer to  $\text{PM}_{10-2.5}$ . However,  $\text{PM}_{2.5}$  may also contain, in addition to the fine-particle mode, some of the lower-size tail of the coarse particle mode between about 1 and 2.5  $\mu\text{m}$   $d_{ae}$ . Conversely, under high relative humidity conditions, the larger particles in the accumulation mode may also extend into the 1 to 3  $\mu\text{m}$   $d_{ae}$  range.
- Three approaches are used to classify particles by size: (1) modes, based on formation mechanisms and the modal structure observed in the atmosphere, e.g., nuclei and accumulation modes which comprise the fine particle mode and the coarse particle mode; (2) cut point, based on the 50% cut point of the specific sampling device, e.g.,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10-2.5}$ , and  $\text{PM}_{10}$ ; and (3) dosimetry, based on the ability of particles to enter certain regions of the respiratory tract.

### 1.2.2 Sources of Airborne Particles in the United States

- The chemical complexity of airborne particles requires that the composition and sources of a large number of primary and secondary components be considered. Major components of fine particles are: sulfate, strong acid, ammonium, nitrate, organic compounds, trace elements (including metals), elemental carbon, and water. Major sources of these fine mode substances are fossil fuel combustion by electric utilities, industry and motor vehicles; vegetation burning; and the smelting or other processing of metals.
- Sulfur dioxide ( $\text{SO}_2$ ), nitrogen oxides ( $\text{NO}_x$ ), and certain organic compounds are major precursors of fine secondary PM.  $\text{NO}$  reacts with ozone ( $\text{O}_3$ ) to form  $\text{NO}_2$ .  $\text{SO}_2$  and  $\text{NO}_2$  react with hydroxy radical ( $\text{OH}$ ) during the daytime to form sulfuric and nitric acid. During the nighttime  $\text{NO}_2$  reacts with ozone and forms nitric acid through a sequence of reactions involving the nitrate radical ( $\text{NO}_3$ ). These acids may react further with ammonia to form ammonium sulfates and nitrates. Some types of higher molecular weight organic compounds react with  $\text{OH}$  radicals, and olefinic compounds also react with ozone, to form oxygenated organic compounds which can condense onto existing particles.  $\text{SO}_2$  also dissolves in cloud and fog droplets where it may react with dissolved  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ , or, if catalyzed by certain

metals, with  $O_2$ , yielding sulfuric acid or sulfates, that lead to PM when the droplet evaporates.

- The formation of secondary PM depends on reactions involving OH,  $O_3$ , and  $H_2O_2$ , species which are normally present in the atmosphere but which are generated in higher concentrations during the photochemical smog formation process. Since smog formation increases with sunlight and temperature, secondary PM peaks during the summer in most U.S. areas.
- Background geogenic and biogenic emission sources include: wind blown dust from erosion and reentrainment; the long-range transport of dust from the Sahara desert; sea salt; particles formed from the oxidation of sulfur compounds emitted from oceans and wetlands; the oxidation of  $NO_x$  from natural forest fires and lightning; and the oxidation of hydrocarbons (such as terpenes) emitted by vegetation.
- Major components of coarse particles are aluminosilicates and other oxides of crustal elements (e.g., Fe, Ca, etc.) in soil dust; fugitive dust from roads, industry, agriculture, construction and demolition; fly ash from combustion of oil and coal; and additional contributions from plant and animal material.
- Fugitive dust constitutes about 90% of estimated  $PM_{10}$  emissions in the United States. Emissions are sporadic and widespread. Only a small percentage of this material is emitted in the fine particle size fraction.
- Uncertainties in emissions inventory estimates could range from about 10% for well defined sources (e.g., for  $SO_2$ ) to an order of magnitude for widespread and sporadic sources (e.g., fugitive dust).
- There has been no clear trend in estimated emissions of fugitive dust and emissions from natural sources from 1984 to 1993. Estimated primary  $PM_{10}$  emissions from combustion sources have decreased by about 10%; estimated  $SO_2$  emissions have decreased by about 6%; and there was no significant change in estimated  $NO_x$  emissions from 1984 to 1993.
- Receptor modeling has proven to be a useful method for identifying contributions of different types of sources especially for the primary components of ambient PM. Apportionment of secondary PM is more difficult because it requires consideration of atmospheric reaction processes and rates. Results from western U.S. sites indicate that fugitive dust, motor vehicles, and wood smoke are the major contributors to ambient PM samples there, while results from eastern U.S. sites indicate that stationary combustion and fugitive dust are major contributors to ambient PM samples in the East. Sulfate and organic carbon are the major secondary components in the East, while nitrates and organic carbon are the major secondary components in the West.
- Fine and coarse particles have distinctly different sources, both natural and anthropogenic. Therefore different control strategies are likely to be needed, depending on whether fine or coarse particles (or both) are selected for control.

### 1.2.3 Atmospheric Transport and Fate of Airborne Particles

- Dry deposition of fine particles is slow. Nuclei-mode (ultrafine) particles are rapidly removed by coagulation into accumulation-mode particles. Accumulation-mode particles are removed from the atmosphere primarily by forming cloud droplets and falling out in raindrops. Coarse particles are removed mainly by gravitational settling and inertial impaction.
- Primary and secondary fine particles have long lifetimes in the atmosphere (days to weeks) and travel long distances (hundreds to thousands of kilometers). They tend to be uniformly distributed over urban areas and larger regions, especially in the eastern United States. As a result, they are not easily traced back to their individual sources.
- Coarse particles normally have shorter lifetimes (minutes to hours) and only travel short distances (<10's of km). Therefore, coarse particles tend to be unevenly distributed across urban areas and tend to have more localized effects than fine particles. (Dust storms occasionally cause long range transport of the smaller coarse-mode particles.)

### 1.2.4 Airborne Particle Measurement Methods

- Measurement of ambient PM mass and chemical composition is important for: source attribution; inventories of the observed mass; health and welfare effects studies; and determination of compliance with standards. A comprehensive approach requires a combination of analytical techniques to assess: (1) mass; (2) elemental composition; (3) water-soluble ionic species; and (4) organic compounds.
- Various sampling systems based on gravimetric (weight) measurements of collected particles yield direct measurements of airborne particle mass. The high volume (hi-vol) sampler, used extensively in the United States before establishment of PM<sub>10</sub> as the indicator for the PM standard, collects and measures the mass of total suspended particulates (TSP), including both fine and coarse particles. Certain other samplers (e.g., dichotomous samplers or impactors) use one or more sampler heads or other separator devices to selectively collect and measure the mass of various size fractions of PM.
- There are no calibration standards for suspended particle mass; therefore, the accuracy of particle mass measurements cannot be determined. The precision of particle mass measurements can be estimated by comparing results from collocated samplers. When using different measurement techniques, samplers of different design or manufacturer, and, in some cases, when using identical systems of different age or cleanliness, substantial biases of 50% or more have been observed. Mass concentration measurements with a precision close to 10% have been obtained with collocated samplers of identical design and same time since cleaning.
- Available technology allows accurate ( $\pm 10$  to 15%) measurement of several of the major components of coarse and fine particles (minerals, sulfates, strong acids, and ammonium). However, collection and measurement technologies for elemental carbon, organic carbon, and nitrates are not as well established.

- Field studies of EPA Equivalent PM<sub>10</sub> Reference Methods and reviews of field data from collocated PM<sub>10</sub> samplers show substantial biases under certain conditions. These biases result from: (a) allowing a tolerance of  $\pm 0.5 \mu\text{m}$  for the  $10 \mu\text{m}$  cutpoint; (b) inadequate restrictions on internal particle bounce; (c) soiling of certain types of PM<sub>10</sub> inlets; and the losses of semivolatile components.
- Semivolatile organic compounds and semivolatile ammonium compounds (such as  $\text{NH}_4\text{NO}_3$ ) may be lost by volatilization during sampling. Such losses may be very important in woodsmoke impacted areas for organic compounds or in agricultural and other areas where low sulfate and high ammonia lead to high  $\text{NH}_4\text{NO}_3$  concentrations.
- Beta attenuation, tapered element oscillating microbalance (TEOM), and optical monitoring methods have been extensively field tested. Although acceptable comparisons with EPA reference sampling methods have been reported in some collocated field studies, significant losses of semivolatile components may occur during and after sample collection. The presence of significant amounts of semivolatile particles at sampling locations in the western United States is a major concern.
- Certain older optical methods, which provided estimates of ambient PM levels used in epidemiology studies of the 1950s to 1970s, are still employed in some countries. These include (a) the black smoke (BS) method, based on light reflectance from particle stains on sample collection filters and extensively used in Britain and elsewhere in Europe; and (b) the coefficient of haze (COH) method, based on light transmission through the filter stain and used in some U.S. areas. Neither method directly measures the mass of collected particles; so credible estimates of particle concentrations (in  $\mu\text{g}/\text{m}^3$ ) can only be made via site-specific calibration against mass measurements from collocated gravimetric sampling devices. BS and COH sampling devices typically have  $\approx 4.5 \mu\text{m}$  cut points, collect mainly fine particles but also some coarse particles up to  $\approx 10 \mu\text{m}$ , and are more comparable to PM<sub>2.5</sub> than PM<sub>10</sub> or TSP measurements. BS and COH readings are especially sensitive to elemental carbon particle concentrations.
- Personal PM exposure samplers are desirable for evaluating individual exposures. Relatively unobtrusive personal samplers have been designed for several particle size cutpoints, and recent studies suggest that acceptable precision is possible, covering the size range from at least  $0.1$  to  $10 \mu\text{m}$  d<sub>ae</sub>.
- Physical elemental analysis methods for metals and other elements include x-ray fluorescence (XRF), particle-induced x-ray emission (PIXE), and instrumental neutron activation analysis (INAA). Atomic absorption spectrometry (AAS) is used for soluble ions such as sodium, magnesium, potassium, and calcium. Ion chromatography (IC) is used for nitrate and sulfate. Automated colorimetry (AC) is used to measure ammonium, chloride, nitrate, and sulfate.
- Accurate chemical speciation of organics, nitrates, and acidity requires comprehensive sampling system components, including gas stream denuders and sequential filter packs. Sampling artifacts can cause significant errors in measurement of organic PM. Some disagreement exists, however, about whether adsorption or volatilization artifacts are most important. Sampling artifacts may be introduced by changes in temperature or organic vapor

concentration during sampling and/or storage. Organic aerosol sampling artifacts can cause large errors in particle mass measurements in areas where a large fraction of the PM mass is organic.

- Chemical analysis of the organic fraction of airborne PM is very costly and difficult because of the complex mixture of hundreds of compounds. Analyses of organic compounds have utilized high-performance liquid chromatography (HPLC) and gas-chromatography combined with mass spectroscopy (GC/MS), but only 10 to 20% of the organic mass has been identified as specific chemical compounds. It is now known that some organic vapors are adsorbed on quartz-fiber filters and that some semivolatile material evaporates during and after sampling. However, most information on organic, elemental, and carbonate carbon comes from samples collected on quartz-fiber filters.
- A variety of methods are now available for sampling or analysis of all types of bioaerosols, including fungal spores, bacteria, pollen, and plant or animal fragments. Analytical methods include: cell culture, microscopy, immunoassay, other bioassay methods, chemical assays, and molecular techniques for DNA/RNA-containing particles.
- Light and electron-microscopy analysis of particle morphology (size and shape) can also be used to help identify sources and transport mechanisms for airborne particles.

### **1.2.5 Ambient U.S. PM Concentrations: Regional Patterns and Trends**

- Particle mass data have been collected at a number of rural, suburban, and urban sites across the United States by various local, state, and national programs. The data have been stored in the Aerometric Information Retrieval System (AIRS). Data have also been collected at remote sites as part of the IMPROVE and NESCAUM networks.
- Estimates of annual average biogenic and geogenic PM<sub>10</sub> concentrations range from 5 to 11  $\mu\text{g}/\text{m}^3$  for the eastern United States and 4 to 8  $\mu\text{g}/\text{m}^3$  for the western United States. Annual average PM<sub>10</sub> concentrations in national parks, wilderness areas, and national monuments in the western United States range from 5 to 10  $\mu\text{g}/\text{m}^3$  (based on data from IMPROVE). The lowest values in AIRS, obtained at remote sites, range from 4 to 10  $\mu\text{g}/\text{m}^3$ . Annual average PM<sub>10</sub> values representative of relatively clean suburban and rural areas reported in AIRS for 1993 ranged from 9 to 13  $\mu\text{g}/\text{m}^3$ .
- The five cities with the highest annual mean PM<sub>10</sub> concentrations for urban sites in the western United States, found in AIRS from 1990 to 1994, were Southern California cities in agricultural regions: Visalia, CA; Bakersfield, CA; Fresno, CA; Riverside, CA; and Stockton, CA. The average concentration in these five areas ranged from 44.8 to 60.4  $\mu\text{g}/\text{m}^3$ .
- Annual average PM<sub>10</sub> concentrations for most urban areas in the United States are typically greater than about 20  $\mu\text{g}/\text{m}^3$ . Highest annual mean PM<sub>10</sub> concentrations in the western United States are significantly higher than corresponding five year annual mean values of about 34  $\mu\text{g}/\text{m}^3$  in eastern U.S. urban areas (Atlanta, GA; Paterson, NJ; Roanoke, VA; Philadelphia, PA; and Atlantic City, NJ) and 36  $\mu\text{g}/\text{m}^3$  in central U.S. urban areas (St. Joseph, MO; Steubenville, OH; Cleveland, OH; Omaha, NE; and Chattanooga, TN). The lowest annual mean PM<sub>10</sub> concentrations found at sites in U.S. populated areas (Penobscot Co., ME;

Marquette, MI; and Lakeport, CA) averaged about  $12 \mu\text{g}/\text{m}^3$  during the period from 1990 to 1994.

- $\text{PM}_{10}$  mass concentrations averaged over regions or by city, using sites in operation during 1988 to 1994, show a significant year to year decrease at most sites. Exceptions are Philadelphia and some locations in Southern California. The regional decreases at urban sites between 1988 and 1994 were: 20% for the contiguous United States; 18% for the eastern United States; and 28% for the western United States.
- Information on trends of  $\text{PM}_{2.5}$  (fine) and  $\text{PM}_{10-2.5}$  (coarse) have been examined. However, the data from Philadelphia, several AIRS sites, the Harvard Six-City sites, and California sites is generally not sufficient either in number of sites or number of years (2.5 to 10 years per site) to demonstrate differential trends in coarse PM and fine PM.
- Long time series for  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  are available from a relatively few sites in Philadelphia and California. Typically,  $\text{PM}_{2.5}$  to  $\text{PM}_{10}$  ratios are greater than 0.5 at these sites on an annually averaged basis. However, values of the ratio of  $\text{PM}_{2.5}$  to  $\text{PM}_{10}$  are highly variable and can be much smaller than 0.5 on individual days. Because of these considerations, values of  $\text{PM}_{2.5}$  should be inferred from  $\text{PM}_{10}$  only where some site-specific information is available. Seasonal or yearly estimates will be more reliable than daily estimates.
- Sulfate ( $\text{SO}_4^{2-}$ ) and strong acidity ( $\text{H}_2\text{SO}_4$  plus  $\text{HSO}_4^-$ ) are regional pollutants distributed relatively evenly over areas of the eastern United States during the summertime. However, in high density livestock areas and the centers of large urban areas, ammonia neutralizes part of the acidity.
- Data for assessing day-to-day variability in  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  are only available from one site in Philadelphia. These data can be used to indicate the potential for daily changes in 24-hour average  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  levels for risk analyses. During this study, average day-to-differences in  $\text{PM}_{2.5}$  were  $6.8 \pm 6.5 \mu\text{g}/\text{m}^3$  and  $8.6 \pm 7.5 \mu\text{g}/\text{m}^3$  for  $\text{PM}_{10}$ . Maximum day-to-day differences were  $54.7 \mu\text{g}/\text{m}^3$  for  $\text{PM}_{2.5}$  and 50.4 for  $\text{PM}_{10}$ .

### 1.2.6 Human PM Exposure

- The total personal exposure to PM consists of outdoor (ambient) and indoor exposures. Nonambient conditions, mainly indoors at home or at work, occupy the vast majority of a person's time. In the U.S., the average daily time spent indoors is 20 h/day, or 85% of the day. Some additional time, about 1.0 to 2.0 h (5%) of the day, is also spent in other nonambient microenvironments (e.g., in vehicles in transit)
- $\text{PM}_{10}$  in ambient air penetrates into residential microenvironments and reaches an equilibrium approaching outdoor concentrations. Once indoors, PM of ambient origin decreases due to deposition on surfaces through gravitational settling and electrostatic attraction. The coarse PM has a much higher deposition rate than the fine PM.
- Human indoor activity (e.g., walking on carpets) tends to resuspend previously deposited  $\text{PM} > 5 \mu\text{m}$  and to stir up or suspend other material (such as tracked-in soil and a variety of



biological material such as mold spores and insect debris). Such activity increases indoor  $PM_{10}$  but not indoor  $PM_{2.5}$ .

- In residential and occupational indoor microenvironments, PM is generated by indoor sources (cooking, smoking, vacuuming, dusting, etc.) and is also removed from the indoor air by gravitational deposition and mechanical means (air cleaners, filters, etc.).
- When a cross-sectional analysis is performed that compares ambient  $PM_{10}$  to individual personal exposures to  $PM_{10}$  for a group of subjects, the correlation often goes towards zero, because of the large influences of indoor sources and sinks that vary *between* the individuals. However, other types of analyses, as follow, indicate significant contributions of ambient particles to total human exposure.
- Because of the relative day-to-day consistency *within* any given residence for the indoor sources and sinks of  $PM_{10}$ , the longitudinal (time-series) correlation of personal exposure of a specific individual to total indoor  $PM_{10}$  (from outdoor *and* indoor sources) and ambient  $PM_{10}$  can be very high. Two analyses conducted to date on a limited number of subjects have yielded  $R^2$  values above 0.9 when indoor sources are consistent from day-to-day.
- Experimental data on a cohort of elderly housewives (N=5) and retirees (N=2), purposefully chosen to have minimal sources of PM at home, shows that their personal exposures to  $PM_{10}$  are highly correlated both with the ambient  $PM_{10}$  immediately outside their homes ( $0.77 < r < 0.96$ ) and at a nearby monitoring station ( $0.75 < r < 0.96$ ). For the identical cohort of elderly housewives and retirees, their personal exposures to  $PM > 10 \mu m$  (TSP -  $PM_{10}$ ) had virtually no correlation with the ambient  $PM > 10 \mu m$  ( $r = -0.03$ ;  $R^2 = 0.00$ ).
- Experimental data on personal exposures to sulfates, which are predominantly of outdoor origin and submicron size, show consistently high correlations of total personal exposures with ambient sulfate ( $0.78 < R^2 < 0.92$ ).
- Both fine and coarse particles will enter homes and other microenvironments. However, because coarse particles are rapidly removed, the indoor concentration of ambient fine particles is more representative of the outdoor or ambient fine particle levels. Because fine particles are evenly distributed and remain suspended indoors, a centrally located ambient monitor may provide a fine-particle concentration that is representative of the community exposure to ambient fine particles, but this will usually not be the case for coarse particles.
- Because indoor sources typically do not generate fine mode particles of the same chemical composition as the most common fine particles of ambient origin (e.g., sulfates, metals, etc.), the presence of indoor PM sources will not change the relationships noted in the immediately preceding bullet. Also, the production of indoor-generated particles is controlled by daily indoor activities. Therefore, the exposure to indoor-generated particles will not be correlated with the concentration of ambient (outdoor-generated) particles, and time-series epidemiology based on ambient measurements will not identify health effects of indoor-generated particles.

- Therefore, the measurements of *daily* variations of *ambient* PM concentrations, as used in the *time-series* epidemiology studies of Chapter 12, have a plausible linkage to the daily variations of human exposures to PM from *ambient* sources, for the populations represented by the ambient monitoring stations. This linkage should be better for indicators of fine particles (PM<sub>2.5</sub>) than for indicators of fine plus coarse particles (PM<sub>10</sub> or TSP), which, in turn, should be better than indicators of coarse particles (PM<sub>10</sub>-PM<sub>2.5</sub>).

### 1.3 DOSIMETRY

For risk assessment purposes, exposure-dose-response models are useful in examining the effects of different host characteristics, activity patterns, and exposures on biological responses. Development of a comprehensive biologically based exposure-dose-response model to aid risk assessment requires more information on mechanisms of action of particles on respiratory tract tissues, including behavior of particles once inhaled or deposited (e.g., hygroscopic growth, disaggregation of aggregated particles), pathological processes affecting deposition and clearance of particles, and factors which influence the response of tissue to particle burden. Deposition and clearance models are useful in estimating average regional particle deposition and regional clearance as a function of major particle characteristics. Dosimetry models have also been useful to characterize average PM deposition patterns for humans as a function of age, gender, and activity pattern and may also ultimately be useful for extrapolating laboratory animal data to estimate concentrations that might be expected to be associated with effects in humans.

- Particles may be deposited in (a) the extrathoracic airways (i.e., mouth, nose, and larynx); (b) in airways of the tracheobronchial region; and (c) in the alveolar region where gas exchange occurs. There are differences in deposition mechanism and dose distribution in each of these areas that are dependent on particle size and airway geometry. The major mechanisms of particle deposition on airway surfaces in the respiratory tract are impaction, sedimentation, diffusion, interception, and electrostatic precipitation.
- Respiratory tract deposition patterns are primarily dependent on particle size and distribution (as indicated by the mass median aerodynamic diameter and the geometric standard deviation) within the inspired air. Biologic effects may be a function not only of particle mass deposition but also particle number or the total surface area of the particles.
- Various host factors have been shown to influence predicted particle deposition patterns including age, ventilation pattern, and the presence of obstructive or inflammatory airway disease. Higher overall ventilation increases total deposition. Increased mouth breathing increases the deposition of coarse particles in the tracheobronchial region. Obstructive airway disease, such as asthma, emphysema, and chronic bronchitis, results in increased deposition of particles in the lower respiratory tract.
- Acute effects of PM are probably best related to deposited dose, whereas chronic effects may be related to cumulative or retained dose. Retention of particles is a function of deposition site, clearance of particles by macrophages or the mucociliary system, and particle

characteristics, especially solubility. Chronic effects may also arise from recurring cycles of pulmonary injury and repair.

- There are substantial differences among laboratory animal species with regard to the inhalability of different sized particles as well as quantitative and qualitative differences in airway geometry.
- With regard to PM, extrapolation of responses from laboratory animals to humans is hampered by limited development of models due to the lack of data characterizing differences in inhalability, airway geometry, and clearance mechanisms among species. In humans, some inhalable particles can exceed  $15\ \mu\text{m d}_{\text{ae}}$ , while in small laboratory animals, inhalable particles seldom exceed  $4\ \mu\text{m d}_{\text{ae}}$ .
- Respiratory tract dosimetry supports the choice of  $\text{PM}_{10}$  as an index of thoracic particles. However, dosimetric considerations do not provide insight into the selection of a size cut to characterize a fine particle mode.

## 1.4 PARTICULATE MATTER HEALTH EFFECTS

Many epidemiologic studies have shown statistically significant associations of ambient PM levels with a variety of human health endpoints, including mortality, hospital admissions, respiratory symptoms and illness measured in community surveys, and changes in pulmonary mechanical function. Associations of both short-term (usually days) and long-term (usually years) PM exposure with most of these endpoints have been consistently observed. The general internal consistency of the epidemiologic data base enhances the confidence accorded the reported results and has contributed to increasing public health concern. However, there remains uncertainty regarding the shapes of PM exposure-response relationships; the magnitude and variability of risk estimates for PM; the ability to attribute observed health effects to specific PM constituents; the time intervals over which PM health effects (e.g., shortening of life) are manifested; the extent to which findings in one location can be generalized to other locations; and the nature and magnitude of the overall public health risk imposed by ambient PM exposure. While the epidemiology data provide support for the associations mentioned above, understanding of underlying biologic mechanisms has not yet emerged.

### 1.4.1 Epidemiology Findings

The findings from the epidemiology studies are often expressed in terms of relative risk (RR), indicating the ratio of the probability of occurrence of a given effect between two different exposure conditions or exposure groups, or as an odds ratio, which is similar to RR for conditions that occur relatively infrequently (such as PM-mortality). Relative risks are often expressed for a specific increase in a PM indicator (e.g., a  $50\ \mu\text{g}/\text{m}^3$  increase in  $\text{PM}_{10}$ ) and provide an estimate of percentage increase in risk above baseline mortality or morbidity rates in the lowest exposure time periods or location per the stated increment of PM indicator concentration. For example, a  $\text{RR} = 1.05$  per  $50\ \mu\text{g}/\text{m}^3\ \text{PM}_{10}$  increase implies that an approximate 5% increase over background risk level is associated with a  $50\ \mu\text{g}/\text{m}^3$  increase in

PM<sub>10</sub> over baseline in the particular study area, assuming linearity of dose-response relationships and the absence of a threshold.

### ***Ambient PM Mortality Effects***

- Early studies of severe air pollution episodes in Europe and the U.S. from the 1930's to 1950's indicated that exposure to high ambient levels of urban air pollution can produce marked increases above the expected numbers of daily deaths during severe episodes (e.g., in the Meuse Valley in 1930, in Donora in 1948, and in London in 1952). These observations left little doubt that exposures to ambient air containing high concentrations of particles and other copollutants such as SO<sub>2</sub> can be lethal, although underlying mechanisms have not yet been delineated.
- More than 20 time-series analyses published in the past 10 years demonstrate positive associations between daily mortality and 24-h concentrations of ambient particles indexed by various measures (BS, COH, TSP, PM<sub>10</sub>, PM<sub>2.5</sub>, H<sup>+</sup>, SO<sub>4</sub><sup>-</sup>) in numerous metropolitan areas of the U.S. and other countries. Relative risk (RR) estimates for daily mortality in relation to daily ambient PM concentration are generally positive and statistically significant across a variety of statistical modeling approaches and methods of adjustment for effects of relevant covariates such as season, weather, and co-pollutants.
- Numerous time-series analyses of TSP-mortality relationships have explored many methodological issues related to use of specific types of models (e.g., parametric, non-parametric, Poisson, GLM, LOESS, etc.), model specification (e.g., inclusion of only PM in analytical models or other copollutants as well), control for impacts of weather variables (temperature, humidity, synoptic weather patterns), and adjustments for other potentially confounding covariates. Several analyses of data from Philadelphia by various investigators have proven to be especially useful in confirming significant positive relationships between 24-h TSP concentrations and daily mortality, while also clarifying season-specific variations in the PM-mortality RR and the impacts of weather adjustments or other copollutants on the RR attributed to PM. Recent Health Effects Institute-sponsored analyses underscore the great complexity inherent in simultaneous statistical adjustment for health effects of multiple air pollutants. Overall, the analyses have produced basically robust results indicative of significant PM effects on mortality.
- RR estimates for total non-accidental mortality associated with a 50 µg/m<sup>3</sup> increase in 24-h average PM<sub>10</sub> range from 1.015 to 1.085. With PM<sub>10</sub> as the only pollutant index in the model, RR = 1.025 to 1.085. In the studies testing multiple pollutant models (with copollutant(s) in the model), PM<sub>10</sub> RR = 1.015 to 1.025. Higher relative risks are indicated for the elderly and for those with pre-existing respiratory conditions.
- The new time-series analyses clearly substantiate significant associations between daily mortality or morbidity and ambient 24-h PM<sub>10</sub> concentrations typical of U.S. urban airsheds. Less extensive evidence points toward fine particles as likely being important contributors to the observed PM-associated mortality, based on studies showing positive associations of daily mortality with various fine particle indicators (e.g., PM<sub>2.5</sub>, SO<sub>4</sub><sup>-</sup>, H<sup>+</sup>, etc.).

- Fine particles (PM<sub>2.5</sub>) showed a consistent and statistically significant relationship to acute mortality in six U.S. cities, with relative risks ranging from 1.02 to 1.06 per 25  $\mu\text{g}/\text{m}^3$  PM<sub>2.5</sub>. Coarse particles showed no significant relationship to excess mortality in five of the six cities. In Steubenville, PM<sub>15-2.5</sub> was more strongly related to mortality than was PM<sub>2.5</sub>, but these two particle indicators were highly correlated.
- Recent chronic (annual average) PM exposure studies also indicate statistically significant positive associations between excess mortality and fine particle indicators. Relative risk estimates derived from such studies suggest greater percentage increases in mortality risk than do the short-term mortality RR estimates. However, the chronic exposure RR estimates are based on PM concentration during the 5 or 15 to 20 year study periods and do not necessarily reflect the full impacts of longer past PM exposures (likely much higher in the most highly polluted cities). Thus, lower RR estimates than the published ones for mortality associated with chronic PM exposure are apt to apply.

### ***Ambient PM Morbidity Effects***

Numerous epidemiologic studies in the United States and elsewhere have also demonstrated significant associations between ambient PM exposures indexed by a variety of indicators (BS, TSP, PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>4</sub><sup>-</sup>, H<sup>+</sup>) and various acute and chronic morbidity outcomes. Such outcomes include, for example, hospital admissions, increased respiratory symptoms, and decreased lung function.

- Studies of hospitalization for chronic obstructive pulmonary disease (COPD) and pneumonia show moderate, but statistically significant RR's in the range of 1.06 to 1.25 per increase of 50  $\mu\text{g}/\text{m}^3$  in PM<sub>10</sub> (24-h). Most studies of hospitalizations for respiratory illnesses typically include patients  $\geq 65$  years of age. Increased hospital admissions for respiratory causes during London episodes suggested an association with sulfuric acid aerosols, as well as with BS and SO<sub>2</sub> levels. Recent studies also show associations between ambient acidic aerosols and summertime respiratory hospital admissions.
- Studies of PM associations with lower respiratory disease yielded odds ratios (OR) which ranged from 1.10 to 1.28, and studies of cough yielded odds ratios ranging from 0.98 to 1.29 for a 50  $\mu\text{g}/\text{m}^3$  increase in PM<sub>10</sub> (24-h). Limited data were available relating PM exposure to asthma or respiratory symptoms in adults. Chronic cough, chest illness, and bronchitis showed positive associations with annual average PM concentrations.
- Pulmonary function studies of children suggest that short term effects result from PM exposure. Peak expiratory flow rates were decreased 30 to 40 ml/sec per 50  $\mu\text{g}/\text{m}^3$  increase in PM<sub>10</sub> (24-h). Somewhat larger effects occurred in symptomatic groups, such as asthmatics. An estimate of the effect of PM on lung function in adults found a 29 ( $\pm 10$ ) ml decrease in FEV<sub>1</sub> per 50  $\mu\text{g}/\text{m}^3$  increase in PM<sub>10</sub>, similar in magnitude to changes found in children. The chronic pulmonary function studies are less numerous than the acute studies and the results are inconclusive.
- Bronchitis symptoms and prevalence rates in children were found to be somewhat more closely associated with annual average H<sup>+</sup> concentrations than with other PM indicators.

However, in studies demonstrating these effects, the acid levels were highly correlated with other fine-particle indicators.

- While numerous studies of PM related respiratory morbidity have been conducted using PM<sub>10</sub> as an indicator, only a few studies have examined the effects of fine particle indicators, e.g., PM<sub>2.5</sub>. Although different studies have suggested that one of these exposure indicators is a better predictor than the other for a specific endpoint, this issue is as yet unresolved. The PM<sub>2.5</sub> studies do show effects related to exposure to the fine fraction, but high correlations among PM<sub>2.5</sub>, PM<sub>10</sub>, and acid aerosols make it very difficult to attribute the effects to a single specific exposure indicator.

### 1.4.2 Toxicology Findings

The data on the potential toxicity of PM are derived from controlled human exposure, laboratory animal, and occupational studies. Each type of study has its own strengths and weaknesses. The studies vary in the populations examined (i.e., by age, health status, etc.), the duration of the study, and the exposure atmospheres (i.e., size distribution, concentration, chemical composition). The studies indicate that inhaled PM causes effects on the respiratory tract. The magnitude and character of the effects are dependent on the particle size distribution and exposure dose, as well as on the physiologic status of the host.

#### *Acidic Aerosols*

Most of the toxicology data on PM are derived from controlled exposure studies in humans and laboratory animals. These studies have most extensively focused on acidic aerosols, namely sulfuric acid aerosol and various sulfates and nitrates, and have included characterization of acid aerosol effects on pulmonary mechanical function, lung particle clearance mechanisms, and other lung defense mechanisms.

- Healthy subjects experience few, if any, decrements in lung function or altered airway responsiveness following single exposures to inhaled acid aerosols (H<sub>2</sub>SO<sub>4</sub>) at levels up to 2,000 µg/m<sup>3</sup> for 1 h. Mild lower respiratory symptoms (such as cough) occur at exposure concentrations in the >500 µg/m<sup>3</sup> range.
- A substantial portion of inhaled acid aerosols may be neutralized by airway ammonia or buffered by airway surface liquids.
- Acid aerosol exposures (>100 µg/m<sup>3</sup>) can cause changes in mucociliary clearance, in healthy or asthmatic humans. Mucociliary clearance in laboratory animals is initially increased and then ultimately decreased by repeated exposures to 125 µg/m<sup>3</sup> H<sub>2</sub>SO<sub>4</sub> aerosol. Chronic exposure of laboratory animals to higher acid levels (~ 250 µg/m<sup>3</sup>) for 52 weeks alters clearance and is also associated with changes in the bronchial tree indicative of mucus hypersecretion.
- Asthmatic subjects are more sensitive than healthy subjects to the effects of acid aerosols on lung function. Responses in asthmatics are generally observed with acute (<3 h) exposures at concentrations of ~ 350 µg/m<sup>3</sup> and higher. Exposures in the 450 to 1000 µg/m<sup>3</sup> range in asthmatics can result in changes in airway responsiveness to bronchoconstrictor agents.

- Adolescent asthmatics may be more sensitive than adults and may experience small decrements in pulmonary mechanical function in response to acute exposures (<3 h) to H<sub>2</sub>SO<sub>4</sub> at levels between 70 and 100 µg/m<sup>3</sup>.
- Acute exposure (<24 h) of healthy laboratory animals to H<sub>2</sub>SO<sub>4</sub> at concentrations above 1000 µg/m<sup>3</sup> can alter pulmonary mechanical function largely due to bronchoconstriction. In guinea pigs, 100 µg/m<sup>3</sup> of acid aerosol may produce small transient effects. Chronic exposure (weeks/months) to 500 µg/m<sup>3</sup> H<sub>2</sub>SO<sub>4</sub> is also associated with alterations in pulmonary mechanical function.
- Submicron droplets of H<sub>2</sub>SO<sub>4</sub> are effective in altering pulmonary mechanical function in laboratory animals. Aerosols larger than 2 to 4 µm have a low inhalability in small laboratory animals (e.g., mice, rats, etc.) However, acid aerosol studies in humans do not permit a clear distinction between responses to aerosols in the range of 0.1 to 20 µm.
- Lung defense mechanisms and resistance to bacterial infection may be altered by exposure to H<sub>2</sub>SO<sub>4</sub> concentrations of about 1000 µg/m<sup>3</sup> in laboratory animal species; alveolar macrophage function may be affected at levels as low as 500 µg/m<sup>3</sup> H<sub>2</sub>SO<sub>4</sub>. Human exposure to acid aerosol (1000 µg/m<sup>3</sup>) did not affect macrophage function.
- Low levels of H<sub>2</sub>SO<sub>4</sub> (100 µg/m<sup>3</sup>) have been shown to react synergistically with O<sub>3</sub>. Exposure of healthy and asthmatic subjects to a mixture of H<sub>2</sub>SO<sub>4</sub> and O<sub>3</sub> suggests that 100 µg/m<sup>3</sup> H<sub>2</sub>SO<sub>4</sub> may slightly exacerbate O<sub>3</sub> lung function effects.
- Acid coating of ultrafine zinc oxide (ZnO) particles appears to enhance the effects of acid on some responses in the guinea pig, including permeability, inflammation, and diffusing capacity. Larger impacts on such endpoints occurred at lower concentrations of H<sub>2</sub>SO<sub>4</sub> and ZnO with combined exposure than with separate exposures to each alone.

### ***Other PM Constituents***

Controlled human exposures to PM constituents other than acid aerosols are limited. Laboratory animal studies and occupational exposure studies provide information on other PM substances, including metals, diesel emissions, crystalline silica, and other miscellaneous particles. Human studies of particles other than acid aerosols provide insufficient data to draw confident conclusions regarding health effects.

- Acute inhalation exposures of humans and laboratory animals to high levels (mg/m<sup>3</sup>) or chronic exposures to lower concentrations of metal particles can have effects on the respiratory tract. The effective exposure levels in such studies are markedly higher than metal concentrations now generally present in the ambient U.S. atmosphere.
- Ultrafine particles occur in the ambient atmosphere in high numbers and have a high collective surface area. The presence of ultrafine particles in human alveolar macrophages suggests human exposure to ambient ultrafines or aggregates of ultrafine particles. Limited human studies indicate slower clearance of ultrafine than of larger inhalable particles. Laboratory animal studies suggest potential toxic effects of inhaled insoluble ultrafine

particles, but the limited available data preclude quantitative estimates of any effective concentrations or doses for ambient ultrafine particle species.

- At very high concentrations ( $>1,000 \mu\text{g}/\text{m}^3$ ) chronic exposures to diesel particles cause inflammatory, histological, and biochemical changes in laboratory animals. The toxicity of diesel emissions is considered to be associated with the particle rather than the gas phase. A no-adverse-effect level for chronic diesel particle exposure has been estimated at  $155 \mu\text{g}/\text{m}^3$ . Thus, at current concentrations ( $< 10 \mu\text{g}/\text{m}^3$ ) typical for U.S. ambient air, diesel PM is not likely to exert significant health effects on healthy humans.
- Chronic exposure to crystalline silica has been shown to cause inflammation of the lung followed by silicosis, a fibrotic lung disease, in occupationally-exposed workers. Using a high estimate of 10% for the crystalline silica fraction in  $\text{PM}_{10}$ , current data suggests that, for healthy individuals not compromised by other respiratory ailments, maintenance of the  $50 \mu\text{g}/\text{m}^3$  annual NAAQS for  $\text{PM}_{10}$  would be adequate to protect against silicotic effects from ambient crystalline silica exposures.

### 1.4.3 Population Groups at Risk

Susceptibility can be affected by factors which influence dosimetry or the response of tissues to particle burdens. The mechanisms by which the various sizes and constituents of ambient PM could exert or modify health effects are not understood. Mechanistic studies to date have mainly focused attention on deposition and clearance mechanisms and less on the biological response to PM. Host factors that may increase the susceptibility to PM include both changes in physiologic factors affecting respiratory tract deposition and pathophysiologic factors affecting response. For example, asthmatics show increased response to acid aerosols or bioaerosols; COPD patients show increased PM deposition and impaired clearance; and airway inflammation or compromised immune status may alter tissue response to inhaled particles.

- Susceptible groups most clearly at special risk for PM effects include the elderly and those with cardiopulmonary disease, based on available epidemiology findings.
- Epidemiology studies indicate that mortality and hospitalization for respiratory causes are strongly related to ambient PM exposures. Several hypotheses have been advanced for possible underlying mechanisms. For example, PM may impair ventilation in COPD patients by causing airway narrowing and increasing the work of breathing. In addition, PM may lead to increased secretion and/or increased viscosity of mucus, possibly exacerbating airway narrowing. Also, some types of PM can cause inflammatory responses and epithelial cell damage in people with chronic respiratory disease.
- Epidemiologic findings indicate that ambient PM exposures are also associated with increased risk for mortality and hospitalization due to cardiovascular causes. Cardiac arrhythmia has been hypothesized as being involved in mortality due to acute PM exposure.
- Epidemiology findings indicate that risk of mortality and morbidity due to lower respiratory disease (e.g. pneumonia) is increased by ambient PM exposure. This may be due to exacerbation, by PM, of already existing respiratory disease. PM may also increase susceptibility to infectious disease by decreasing clearance, impairing macrophage function,



or through other specific and nonspecific effects on the immune system. The epidemiologic findings also indicate that individuals with preexisting infectious respiratory disease (e.g. pneumonia) are at increased risk for PM effects.

- Smokers constitute a significant fraction (ca. 80%) of individuals with COPD and a smaller but notable portion of cardiovascular disease patients. Therefore, smokers are another population group at likely increased risk for PM health effects.
- Asthmatics are more responsive than non-asthmatics to acid aerosols in controlled exposure studies. Asthma exacerbations are well known to be associated with ambient and indoor bioaerosols. In epidemiological studies, asthma exacerbations, sometimes requiring medical attention have also been associated with ambient coarse PM dominated PM<sub>10</sub> exposure.
- Children and adolescents may also be potentially susceptible to ambient PM effects due to their increased ventilatory frequency resulting in greater respiratory tract PM deposition. In children, epidemiologic studies reveal associations of PM exposure with increased bronchitis symptoms and small decreases in lung function.

## **1.5 WELFARE EFFECTS**

Chapter 8 discusses visibility and climate change impacts of airborne particles as two key types of welfare effects associated with ambient airborne particulate matter. Chapter 9 discusses damage to materials due to PM and related pollutants. PM-related effects on vegetation, crops, and ecosystems are not covered in this document.

### 1.5.1 Visibility Effects

- Chapter 8 of this document includes information supplementary to several other significant reviews of the science of visibility, including the 1991 report of the National Acid Precipitation assessment Program, the National Research Council's *Protecting Visibility in National Parks and Wilderness Areas* (1993), and EPA's 1995 *Interim Findings on the Status of Visibility Research*. The following points are made in Chapter 8, and/or in the above referenced documents.
- The relationships between air quality and visibility are well understood. Ambient fine particles are the major cause of visibility impairment. Significant scientific evidence exists showing that reducing fine particle concentrations will improve visibility.
- The National Research Council defines visibility qualitatively as "the degree to which the atmosphere is transparent to visible light." This definition may be expressed quantitatively in terms of contrast transmittance. The EPA has defined visibility impairment as a reduction in visual range and atmospheric discoloration.
- Light, as it passes through the atmosphere from a scene to an observer, is both scattered and absorbed. The rate of loss of transmitted light intensity with distance is measured by the light-extinction coefficient which may be expressed as the sum of the coefficients for: (a) light scattering due to gases; (b) light scattering due to particles; (c) light absorption by gases, and; (d) light absorption by particles.
- Light scattering efficiency depends on particle size, falling off rapidly for particles below 0.3 or above 1.0  $\mu\text{m}$  in diameter. Therefore, particles in the accumulation mode (of the fine particle mode) are most effective in scattering light and are more important in visibility degradation than either nuclei mode or coarse mode particles. Light absorption is not a strong function of particle size. Under exceptional circumstances, such as dust storms, coarse particles can dominate scattering.
- In addition to reducing the intensity of light carrying information about a scene (transmitted radiance), particles also scatter light into the observer's view. This extraneous light, called air light or path radiance, carries no information about the scene. The competition between these two sources of light, expressed as the ratio of transmitted radiance from the scene to path radiance, determines the contrast transmittance and the visual quality of the view.
- Visibility at any location is affected by air quality and non-air quality related effects. The visibility effects of atmospheric constituents are dependant upon not just the mass of pollutants, but on the size distribution and refractive index of particles, which are strongly influenced by relative humidity. Non-air quality effects include the angle between the sun and the observer's sight path, location of clouds, and reflectivity of the ground. These effects are independent of effects due to changes in atmospheric constituents. Lighting and scene effects can be accounted for by defining a range of these effects when estimating visibility changes due to air quality influences.
- The relationship between air pollution and the appearance of a scenic view is well understood. Models exist that, given an adequate description of the air quality and non-air

quality variables, can produce a simulated photograph that accurately depicts a cloud-free scene as it would appear to a human observer.

- There are several potential quantitative indicators of visibility. Indicators such as contrast transmittance, which provide the most information on the appearance of a scenic view, provide little information that is helpful in developing control strategies to improve or protect visibility. Indicators such as fine particle mass and composition provide more information useful for control strategies but provide less information on visibility. Potential indicators include: (a) fine particle mass and composition (fine particle mass alone provides less of both types of information); (b) scattering by dried ambient particles; (c) scattering by particles under ambient conditions; (d) extinction (calculated from measurements of scattering plus absorption); (e) light extinction measured directly; and (f) contrast transmittance.

### **1.5.2 Climate Change**

- Particles suspended in the atmosphere affect the earth's energy budget and thus exert an impact on climate: (a) directly by increasing the reflection of solar radiation by cloud-free portions of the atmosphere, and (b) indirectly by affecting cloud microphysical properties in ways that increase the brightness and stability of clouds.
- Estimates of atmospheric sulfate aerosol solar radiation effects (expressed as radiative forcing) range from  $-0.3 \text{ W m}^{-2}$  to  $-1.1 \text{ W m}^{-2}$  for direct effects and range from  $-0.4$  to  $-1.6 \text{ W m}^{-2}$  for indirect effects. These values may be compared to the estimated radiative forcing of  $+2.4 \text{ W m}^{-2}$  due to the increase in concentrations of greenhouse gases from the pre-industrial era to 1994.
- Therefore, on a globally averaged basis, radiative cooling due to anthropogenic particles may have substantially offset the radiative heating due to increases in atmospheric concentrations of greenhouse gases such as carbon dioxide, methane, and chlorofluorocarbons.
- Aerosol lifetimes are also much shorter than the time required for global mixing, therefore, aerosol radiative effects are most likely to exert their influence on a regional rather than on a global basis.
- The lifetimes of particles in the troposphere are short (days to weeks) compared to the above greenhouse gases (years to over 100 years). Therefore, aerosol concentrations will respond more rapidly to variations in emissions than will the greenhouse gases.

### **1.5.3 Materials Damage**

- Particle exposure results in the soiling of painted surfaces and other building materials, increasing the cleaning frequency for exposed surfaces and possibly reducing their useful lifetimes.
- Evidence suggests possible effects of particles on fabrics, electronics, and works of art.

- Building materials (metals, stones, wood, paints) undergo wear even in the absence of pollutant exposure through physical, chemical, and biological interactions involving moisture, temperature, oxygen, and various types of biological agents.
- Deposition of acid aerosols may increase the corrosion of metals by reacting directly with the metal or the metal corrosion film.
- Deliquescent or hygroscopic salts, deposited on metals, accelerate corrosion by decreasing the critical relative humidity. The decrease in the critical relative humidity results in an increase in the amount of moisture on the metal surface. Acid forming gases dissolve in the moisture layer, causing generation of corrosive acids and ionic species. Exposure to acid forming gases may also limit the life expectancy of paints and may damage various building stones and cement products beyond that resulting from natural weathering processes.
- There are insufficient data available to accurately estimate economic impacts of exposure of building materials to acid forming aerosols and particles.

## 1.6 KEY CONCLUSIONS

- Epidemiologic studies show consistent positive associations of exposure to ambient PM with health effects, including mortality and morbidity. The observed associations of ambient PM exposure with health effects must be adjusted for the effects of other environmental or demographic factors, depending on whether the effects are acute or chronic, in order to quantitatively assess the role that may be attributed to PM exposure. Estimates of PM health effects have shown reasonable quantitative consistency in different studies, with only modest sensitivity to different methods of analysis. However, a clear understanding of specific biologic mechanisms remains to be established.
- Individuals with cardiovascular or pulmonary disease, especially if they are elderly, are more likely to suffer severe health effects (mortality or hospitalization) related to PM exposure than are healthy young adults. Children and asthmatics are also susceptible to certain PM effects, e.g., increased respiratory symptoms and decreased lung function. Smokers also constitute a population group at increased risk for ambient PM exposure effects.
- Recent analyses continue to support the use of  $PM_{10}$  as an indicator of ambient particle exposures associated with human health effects. The consistent association of mortality and various morbidity end points with  $PM_{10}$  exposure clearly substantiates the earlier rationale underlying selection of this indicator and  $PM_{10}$  standard for protection of public health.
- Additional consideration of the subdivision of  $PM_{10}$  into fine and coarse components is also warranted. Indices of PM exposure that have been most consistently associated with health endpoints are by  $PM_{10}$  or  $PM_{15}$  and fine particle indicators. Less consistent relationships have been observed for TSP and the coarse fractions of  $PM_{10-2.5}$ .
- In human populations, the daily variation in the personal exposure to ambient fine particles is reflected by daily variation in ambient fine particle concentration measured at a central

monitoring site better than total exposures to coarse particles are reflected by measurements of ambient coarse particles at community monitoring sites. This is consistent with the observed high correlations of personal sulfate exposures with ambient sulfate concentrations found experimentally. Therefore, central site measurements of fine particle indicators can be useful in PM epidemiology studies.

- Development of a comprehensive biologically-based exposure-dose-response model to aid health risk assessment requires further data characterizing differences in inhalability, airway geometry, and clearance rates among species. Information is also required on mechanism(s) of action, pathological processes affecting deposition and clearance of particles, and factors which influence the response(s) of respiratory tract tissues to particle burden.
- Estimation of public health impacts of ambient airborne particle exposures in the United States would most credibly require use of relative risk estimates derived for particular U.S. urban areas, in combination with estimates of exposures to ambient particle concentrations for the general population and/or specific susceptible subgroups (e.g., the elderly) within those particular areas. In view of geographic differences in ambient PM mixtures and demographics, broad generalization and application of some single "best estimate" of relative risk for a given increment in concentration of a given particle indicator (e.g.,  $PM_{10}$ ,  $PM_{2.5}$ , etc.) would be subject to much uncertainty.
- Epidemiological studies indicate increased health risks associated with exposure to PM, alone or in combination with other air pollutants. PM-related increases in individual health risks are small, but likely significant from an overall public health perspective because of the large numbers of individuals in susceptible risk groups that are exposed to ambient PM.  $PM_{10}$  and indicators of fine particles are more consistently associated with health risks than indicators of coarse particles.
- Aerosol effects on visibility and climate, through light scattering and changes in cloud microphysics, primarily arise from fine particles.
- Based on points discussed above, fine and coarse particles should be considered as separate subclasses of pollutants. Consideration of formation, composition, behavior, exposure relationships, and sources argue for monitoring fine and coarse particles separately. Because fine and coarse particles are derived from different sources, it is also necessary to quantify ambient levels of fine and coarse particles separately in order to plan effective control strategies.